

Investigation of the possibility of surface plasmon polariton excitation on Si surface by femtosecond laser pulses for periodic surface structure formation

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The mechanisms of ripple formation on silicon surface by femtosecond laser pulses are investigated. We demonstrate the transient evolution of the density of the excited free-carriers. As a result, the experimental conditions required for the excitation of surface plasmon polaritons are revealed. The periods of the resulting structures are then investigated as a function of laser parameters, such as the angle of incidence, laser fluence, and polarization. The obtained dependencies provide a way of better control over the properties of the periodic structures induced by femtosecond laser on the surface of a semiconductor material.

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I. INTRODUCTION

Femtosecond lasers are known to be powerful tools for micro- and nanomachining. In particular, these lasers can induce periodic modulations ("Laser-Induced Periodic Surface Structures" or LIPSS) on the surfaces of metals, semiconductors and dielectric samples at relatively moderate laser fluences^{1–6}. Furthermore, it is possible to decrease periods of these structures below the laser wavelength, thus rising the precision of laser nanomachining beyond the diffraction limit^{7–10}. Applications of the ripple structures are numerous. For instance, it is possible to colorize metals and to control over laser marking^{11,12}.

For further development of these applications, it is important to better understand the mechanisms of ripple formation by femtosecond laser pulses. The number of the observed structures is, however, very large making the control over the laser parameters very complicated. In general, two types of structures can be distinguished (i) resonant structures, where the resulting period is correlated with laser wavelength, and (ii) non-resonant structures, which are not explicitly connected with the laser wavelength and with the coherent effects. Thus, the so-called Low-Spatially-Frequency LIPSS (LSFL) are belong with the resonant structures⁸. In addition, much larger parallel ripples of several micrometers, can be considered to belong with the non-resonant structures. In this case, a large number of pulses is required to provide a thick melted depth comparable to the structure amplitude, so that these ripples are rather connected with the capillary wave generation or surface stress⁵. The non-resonant structures were also explained by the self-organized processes, described by the *Kuramoto-Sivashinsky* equation^{13–15}. With the increase in laser pulses and fluence, drop-like structures named "beads", and then conical structures called "black silicon" can be also obtained^{4,16–19}.

In this paper, we focus our attention at the resonant periodic structures with the period near the laser wavelength e.g. the LSFL. The classical theory of ripple formation proposes that scattering of the laser wave by surface roughness couples the laser wave with the surface modes, which interfere with the laser light, and thus lead to a periodic modulation of the absorbed energy²⁰. This theory was recently confirmed by the numerical calculations based on the system of Maxwell equations for rough surface²¹. This scenario requires the presence of an initial roughness of a certain size. Both the laser parameters and the surface roughness are, however, often unknown. In addition, several laser pulses are frequently re-

quired to form a structure. In this case, the period of the energy deposition can be smaller than laser wavelength, as was explained by Bonse et al²² by using the "Sipe-Drude" model. In this way, an explanation of the very narrow structures (HSFL) appearing after numerous pulses^{23,24} was proposed. In addition, Tsibidis et al.²⁵ recently investigated the cumulative hydrodynamic effects and the corresponding surface modifications. It was found that a non-resonant mechanism explains the reduction of the LSFL periodicity with the increase of pulse number in Si. In addition, the possibility to create periodic surface modifications with a single femtosecond pulse was demonstrated for metals and semiconductor materials^{18,22,26}. To explain the formation of the near-wavelength ripples at intense and reduced number of pulses, several authors have proposed the surface plasmon polaritons (SPP) as the mechanism responsible for the surface wave generation in semiconductors and dielectrics^{22,27,28}. The surface plasmon polaritons are known to be excited on metal surfaces. The ripple formation mechanism for metals has been linked with the excitation of surface plasmon polariton by several authors^{29,30}. However, the excitation conditions remain rather puzzling in the case of semiconductor or dielectric materials. Moreover, it is not clear if the SPP coupling can occur by using a single femtosecond laser pulse, since specific coupling conditions are required to add the missing momentum at the surface. In particular, gratings, snom probe, prism, defects, or roughness^{31,32} typically help to couple the laser wave with a surface wave mode. Thus, our study focuses on the semiconductor case, and the analyzed material is monocrystalline Si.

In the case of semiconductor materials, laser-induced modification of the dielectric function changes material properties. As a result, the importance of a transitory metallic state was underlined by Bonse et al³³. In such cases, however, the presence of a nanometric defect (such as bubbles or nanoparticle) at the surface is required and those methods correspond to the well-known case of localized surface plasmon (LSP) excitation around isolated defects³⁴. It was demonstrated, furthermore, that the transient modification of the solid properties follows the plasma dynamics of the free-carrier gas, due to their excitation by the intense laser^{35,36}. However, a systematic study is still required for the conditions of SPP excitation on semiconductor surface by a femtosecond laser interaction. Under these excitation, electron-hole pairs are generated, but also thermal effects play a role. That is why a clear explanation is required for the laser parameter range, ambient environment and sample surface conditions. To help developing the corresponding applications,

the resulting ripple periods should be connected with laser parameters. In this paper, we consider the conditions required to excite surface plasmon polaritons on semiconductor's surface. The developed model provides all the parameters, which lead to the SPP excitation, such as angle of incidence, laser fluence, pulse duration, and surface roughness for the given ambient optical properties. It is demonstrated that, under the required conditions, Si surface becomes optically active under femtosecond irradiation, and thus, SPPs can be excited by irradiation of a coupling device. The resulting periods are analyzed as a function of laser parameters.

The paper is organized as follows. In Section II, we present the experimental protocol. In Section III, we present the model and consider the modification of the optical properties of Si under femtosecond irradiation. In Section IV, conditions of the excitations of the surface plasmon polaritons are presented. In Section V, the laser parameters allowing the SPP excitation are examined as a function of laser fluence and laser pulse duration. Then, the required minimal roughness thickness is discussed. Finally, the calculated periodicities are compared to the experimental values as a function of laser fluence. The evolution of the ripple period is analyzed as a function of the angle of incidence and laser polarization.

II. EXPERIMENTAL DETAILS

The micromachining experiments were performed by using a Ti-sapphire laser (Hurricane model, Spectra-Physics) that was operated at 800 nm, with an energy of $500 \mu J$, a repetition rate of 1 kHz and a laser pulse duration of 100 fs. Laser irradiation of silicon surface was carried out in a vacuum system with a pressure of $5 \cdot 10^{-5}$ to $1 \cdot 10^{-5}$ mbar. This low pressure considerably reduces the redeposition of unwanted debris from the laser ablation process. To get a more uniform laser energy distribution, only the center part of the gaussian laser beam was selected using a square mask of $2 \times 2 \text{ mm}^2$. A spot about $35 \times 35 \mu m^2$ area was obtained projecting the mask image onto the sample surface with a lens ($f' = 50 \text{ mm}$). Laser beam was perpendicular to the sample surface. The laser energy delivered to the sample surface could be attenuated by coupling an analyzer and a polarizer and completed by a set of neutral density filters. The analyzer rotation placed in front of the polarizer are controlled by a computer. The engraving results are in situ monitored by a CCD camera. The number of pulses is controlled by triggering a Pockels

Physical meaning	Notation	Value	Unit	Reference
One-photon absorption coefficient	σ_1	$2\omega Im\sqrt{\varepsilon_\infty(\omega)}/c = 1.021 \cdot 10^5$	m^{-1}	(d, g)
Two-photon ionization rate	σ_2	$0.1 \cdot 10^{-9}$	$m.W^{-1}$	(c)
Impact ionization probability rate	δ_I	$3.6 \cdot 10^{10} e^{-E_g/k_B T_e}$	s^{-1}	(b, e, f)
Auger recombination rate	C	$3.8 \cdot 10^{-43}$	$m^6.s^{-1}$	(b)
Recombination delay at high density	τ_0	6	ps	(a)

TABLE I: The calculation parameters for c-Si under 800 nm irradiation. References: (a) Ref.³⁷, (b) Ref.³⁸, (c) Ref.³⁹, (d) Ref.⁴⁰, (e) Ref.⁴¹, (f) Ref.⁴², (g) Ref.⁴³.

cell, thus reducing the repetition rate of the laser pulse to 5 Hz. We irradiated a <100> monocrystalline silicon (c-Si) wafer by one or several laser pulses at fluences of $0.5 J/cm^2$, $0.8 J/cm^2$, and $1.15 J/cm^2$. Two series of experiments were performed. (i) At very low (one or two) number of pulses, the angle of incidence has been kept normal to the surface. (ii) At $N = 10$ pulses, the angle of incidence and the laser polarization have been varied.

III. MODELING DETAILS

Femtosecond laser can promote carriers from the valence band of a semiconductor to the conduction band leading to free-carrier absorption. In our model, the number density of the carriers in the conduction band is calculated by solving the following equation

$$\frac{\partial n_e}{\partial t} - \nabla \cdot (k_B T_e \mu_e \nabla n_e) = G_e - R_e \quad (1)$$

where n_e denotes the electron-hole pair density, and $G_e = \left[\frac{\sigma_1 I}{\hbar \omega} + \frac{\sigma_2 I^2}{2 \hbar \omega} + \delta_I n_e \right] \frac{n_v}{n_e + n_v}$ is the gain of free-carriers per time unit and volume unit ($m^{-3}.s^{-1}$). n_v is the quantity of valence band electrons, and n_e is the quantity of conduction band electrons.

Both one-photon interband cross-section (σ_1) and the two-photon cross-section (σ_2) are used in the model (Table I). The conduction band can be also populated due to the electron impact ionization (avalanche process). The corresponding coefficient δ_I is also given in Table I. $R_e = \frac{n_e}{\tau_0 + \frac{1}{C n_e^2}}$ is the loss of conduction electrons by Auger recombination, where the recombination time τ_0 is equal to 6 ps in our calculations^{37,44}.

The initial density of free-carriers present in the conduction band is $1.84 \cdot 10^9 cm^{-3}$ at a temperature of 300 K. k_B is the Boltzmann constant. In the near-ablation regime, using

low energy photons (1.5 eV in our case), the number of excitable electrons is limited to the ones available in the valence band. Even in ablation regime at the considered laser intensities, less than one electron per atom is usually promoted to the conduction band⁴⁴. Thus, the number of the excitable valence band electrons is described by $n_0 = \rho_{Si} = 5 \cdot 10^{22} \text{ cm}^{-3}$, equal to the density of the Si lattice. During the excitation, the number of valence band electrons n_v is therefore calculated by $n_v = n_0 - n_e$.

The free-carrier mobility is described by $\mu_e = \frac{e}{m_e \nu}$ where $\nu = 1.5 \cdot 10^{14} \text{ s}^{-1}$ is the free-carrier collision frequency. Collision frequency is adjusted in agreement with melting fluence and melted depth given by Bonse et al⁴⁵, consistent with Monte Carlo simulations of collision frequency in Si⁴⁶. m_e is the optical mass of electron-hole pairs, which is equal to³⁶ $m_e = 0.18m_{e0}$, where m_{e0} is the electron mass. The density of electrons is calculated by using Eq. (1) taking into account thermal diffusion and Auger recombination. The hole temperature and density are considered equal to the ones of free-electrons, since the contribution of the electron-hole pairs to the absorption is taken into account by the optical mass in dielectric function.

Laser energy absorption is calculated as follows^{38,44}

$$\frac{\partial I}{\partial z} = -\alpha_{fcr} I - (\sigma_1 I + \sigma_2 I^2) \frac{n_v}{n_e + n_v}, \quad (2)$$

where I is the local intensity. Intensity at the surface is given by $I_{z=0}(t, x) = [1 - R(x)] I_0(t, x) \frac{1}{\cos \theta}$ and $I_0(t, x) = \frac{2F}{\tau} \sqrt{\frac{\ln(2)}{\pi}} e^{-\frac{1}{2}(\frac{x}{\sigma_x})^2} e^{-\frac{1}{2}(\frac{t-t_0}{\sigma_\tau})^2}$. F denotes the maximum fluence reached during the interaction. $t_0 = 0$ in our calculations. Spot size w_0 and pulse duration τ are respectively defined at the FWHM of spatial and temporal gaussian distributions. Thus, $\sigma_x = \frac{w_0}{2\sqrt{2\ln 2}}$ and $\sigma_\tau = \frac{\tau}{2\sqrt{2\ln 2}}$. The free-carrier absorption is described by $\alpha_{fcr} = \frac{2\omega n_2}{c}$ where $n_2 = \sqrt{0.5 \left(-\Re(\varepsilon_{fcr}) + \sqrt{\Re(\varepsilon_{fcr})^2 + \Im(\varepsilon_{fcr})^2} \right)}$ and $\varepsilon_{fcr} = 1 - \frac{\omega_p^2}{\omega^2} \frac{1}{1+i\frac{\nu}{\omega}}$ describes the dielectric response of the free-carriers.

During the interaction, the surface reflectivity changes depending on the angle of incidence θ and on laser polarization. We take into account the contribution of each layer to the surface reflectivity by using a recurrence formula given by the transmission matrix model⁴⁷⁻⁴⁹

$$R_{i,k} = \left| \frac{r_{i,i+1} + r_{i+1,k} e^{2i\phi_{i+1}}}{1 + r_{i,i+1} r_{i+1,k} e^{2i\phi_{i+1}}} \right|^2,$$

where $\phi_{i+1} = \frac{2\pi h_{i+1}}{\lambda} \sqrt{\varepsilon_{i+1}} \cos \theta_{i+1}$ is the phase shift induced by the $i+1$ layer, h_{i+1} is the

thickness of the layer $i + 1$, and θ_{i+1} is the angle of incidence at layer $i + 1$ given by Snell-Descartes law $\theta_{i+1} = \arcsin \left(\sqrt{\frac{\varepsilon_i}{\varepsilon_{i+1}}} \sin \theta_i \right)$. $r_{i,i+1}$ is given by Fresnel equations⁵⁰. ε_i is the full dielectric function given by Eq. (3) calculated at the layer i , and θ_i is the angle of incidence at the layer i . The dielectric function is calculated as follows^{36,44}.

$$\varepsilon_{Si}(\omega, n_e, \nu) = 1 + (\varepsilon_\infty(\omega) - 1) \frac{n_v}{n_e + n_v} - \frac{\omega_p^2}{\omega^2 (1 + i \frac{\nu}{\omega})} \quad (3)$$

where $\varepsilon_\infty(\omega)$ is the dielectric constant and depends on laser wavelength⁴⁰. n_e is the free-carrier density, ω_p is the plasma pulsation defined here by $\omega_p^2 = \frac{n_e e^2}{m_e \varepsilon_0}$. ω is the laser pulsation defined by $\omega = \frac{2\pi c}{\lambda}$. Photo-emission and thermo-emission are neglected, since Dumber field strongly limits the transport and thus the loss of free-carriers at the surface^{44,49}.

Several parameters of the above model depend on temperature. Under femtosecond laser irradiation, the electron-hole sub-system is excited to much higher temperatures and electron heat conductivity is much larger than that of lattice. To calculate temperatures of carriers and lattice, a two-temperature model is used. The laser-excited zone is small, so that ballistic effects should be also accounted for the electron sub-system. Therefore, we use the following ballistic-diffusive equation⁵¹⁻⁵³ to describe the electron-hole subsystem.

$$\frac{1}{4\nu} \frac{\partial^2 T_e}{\partial t^2} + \frac{\partial T_e}{\partial t} = \nabla (D_{SBD} \nabla T_e) - \frac{\gamma_{ei}}{C_e} (T_e - T_i) + \frac{Q_e}{C_e},$$

where $D_{SBD} = \frac{L^2 \nu}{6\pi^2} [\sqrt{1 + 4\pi^2 K n^2} - 1]$ is the effective free-carrier thermal diffusion term, based on the ratio $Kn = \frac{l_{MFP}}{L}$ of free-carrier mean-free-path $l_{MFP} = \frac{1}{\nu} \sqrt{\frac{3k_B T_e}{m_e}}$ and size of the excited zone $L \sim (2\omega \Im m \sqrt{\varepsilon_{Si}}/c)^{-1}$. Specific heat capacity of free-carriers is taken to be equal to the classical limit $C_e = \frac{3}{2} k_B n_e$. The source term $Q_e = \left[(\hbar\omega - E_g) \frac{\sigma_1 I}{\hbar\omega} + (2\hbar\omega - E_g) \frac{\sigma_2 I^2}{2\hbar\omega} - E_g \delta_I n_e \right] \frac{n_0 - n_e}{n_0} + \alpha_{fcr} I + E_g R_e - \frac{3}{2} k_B T_e \frac{\partial n_e}{\partial t}$ describes the energy of free-carriers by taking into account one-photon and two-photon ionization, the energy loss by electron avalanche, the free-carrier heating, the release of energy due to Auger recombination and last term comes from the variation of the specific heat capacity with time, since density is strongly modified during the pulse.

The time of free-carrier coupling to the lattice was experimentally determined by Sjodin et al³⁹ as a function of free-electron density given by $\tau_\gamma = \tau_{\gamma 0} \left(1 + \frac{n_e}{n_{th}} \right)^2$ with $n_{th} = 6.02 \cdot 10^{20} \text{ cm}^{-3}$ and $\tau_{\gamma 0} = 240 \text{ fs}$. In our calculations, the coupling rate is given by⁴⁴ $\gamma_{ei} = \frac{C_e}{\tau_\gamma}$. Because of the slow thermal diffusion of the lattice energy, we describe the

temperature of the lattice T_{Si} by a classical diffusion equation, taking into account the energy transferred from free-carriers as follows

$$C_{Si} \frac{\partial T_{Si}}{\partial t} = \nabla (\kappa_{Si} \nabla T_{Si}) + \gamma_{ei} (T_e - T_{Si})$$

The specific heat capacity of Si is a function of the liquid density fraction. For solid state, dependence with temperature is given by relations³⁸ $C_{s-Si}[J.m^{-3}] = 10^6 [1.978 + 3.54 \cdot 10^{-4} \cdot T - 3.68 T^{-2}]$ and $\kappa_{s-Si}[W/m/K] = 10^2 [1585 T^{-1.23}]$. For liquid state, parameters are given by⁵⁴⁻⁵⁶ $C_{l-Si}(T) = 1.045 \cdot 10^3 \rho_{l-Si}$, where $\rho_{l-Si} = 2520 \text{ kg/m}^3$ and $\kappa_{l-Si}(T) = 10^2 [0.502 + 29.3 \cdot 10^{-5} (T - T_m)]$. Melting temperature T_m depends on the free-electron density as described by Eq. (4). During the phase transition, both Si heat capacity and conductivity are calculated using the fraction of liquid η , and are respectively defined by $C_{Si}(T) = (1 - \eta) C_{s-Si}(T) + \eta C_{l-Si}(T)$ and $\kappa_{Si}(T) = (1 - \eta) \kappa_{s-Si}(T) + \eta \kappa_{l-Si}(T)$. Melting is considered by using melting enthalpy ΔH_m at the melting temperature T_m . The resulting thermal energy is given by $U = \int_{T_0}^{T_m} C_{s-Si}(T') dT' + \Delta H_m + \int_{T_m}^T C_{l-Si}(T') dT'$ where U is the internal energy of the lattice, T is the Si temperature, $T_0 = 300 \text{ K}$ is the initial temperature of the system, and $\Delta H_m = 4 \cdot 10^9 \text{ J/m}^3$ is the melting enthalpy of Si.

Previously, two different types of phase transitions were shown to take place for Si⁵⁷⁻⁵⁹. The first one is thermal melting and is described by a thermal criterion based on the required energy $E_m = k_B T_m \rho_{Si} + \Delta H_m$. The second phase transition mechanism is a so-called "non-thermal melting" due to the lattice decomposition due to a large number of carriers in the conduction band. The contribution of the non-thermal melting is also taken into account by the decrease of the band gap energy as a function of free-carrier density³⁸ (limited to positive or null values) expressed by $E_g(T, n_e) = 1.17 - 4.73 \cdot 10^{-4} \frac{T^2}{T+636} - 1.5 \cdot 10^{-10} n_e^{1/3}$ and by a lowering of the melting temperature described by the relation⁶⁰

$$T_m = T_m^0 - \frac{n_e E_{gap}}{C_{s-Si}} \quad (4)$$

where $T_m^0 = 1687 \text{ K}$.

Boundary conditions for transport equations are set so that free-carriers do not leave the sample. The sample is $250 \mu\text{m}$ thick, and the optical transmission has been checked to be zero through the sample.

IV. SURFACE PLASMON POLARITONS

Light can be coupled from free space into the surface plasmon polaritons (SPP) only by matching the momentum of the SPPs. This can be done via index matching³¹, or grating coupling^{61,62}. In addition, other cases can be considered. A non-resonant excitation can be performed by scattering of the laser wave on surface defects or a surface roughness. In such a case, laser wave is scattered on a broad angular distribution, and a part of the laser energy couples with surface modes. Laser wave can also interact with near-wavelength structures as described by Mie scattering, which leads to the excitation of localized surface plasmons (LSP)^{34,63}.

In each case, the excitation of surface waves requires several resonance conditions^{31,32,64}. In this part, we present theoretical conditions allowing the excitation of the Surface Plasmon Polaritons (SPPs) at the laser-irradiated surface of Si.

The excitation conditions of surface plasmon polaritons at a flat surface is that the corresponding curves cross in the dispersion diagram^{31,64}. The dispersion relation is obtained from the boundary conditions of the electric and magnetic field at the interface. The continuity of the electric field at the interface results to the expression $\frac{k_2}{k_1} = -\frac{\varepsilon_2}{\varepsilon_1}$, where $\varepsilon_{1,2}$ are the dielectric constants on both sides of the interface, and $k_{1,2}$ are the respecting momenta of the both sides of the interface. In the general case, this expression can be verified only if

$$\Re(\varepsilon_1) \Re(\varepsilon_2) < 0 \quad (5)$$

which corresponds to a metal-dielectric interface. The continuity of the magnetic field at the interface leads to the expression of the SPP wave-number

$$\beta = \frac{\omega}{c} \sqrt{\frac{\varepsilon_1 \varepsilon_2}{\varepsilon_1 + \varepsilon_2}} \quad (6)$$

and to a second SPP excitation condition given by

$$\Re(\varepsilon_2) < -\Re(\varepsilon_1) \quad (7)$$

By introducing Eq. (3) into Eq. (6) and considering an interface with vacuum ($\varepsilon_1 = 1$), we calculate the dispersion relation as a function of the free-carrier density. Figure 1 presents the dispersion relation of Surface Plasmon Polariton and laser wave in the case of Si, as a function of the laser frequency, normalized to plasma frequency. In this calculation, the band structure of Si has been taken into account by replacing $\varepsilon_\infty(\omega)$ in Eq.

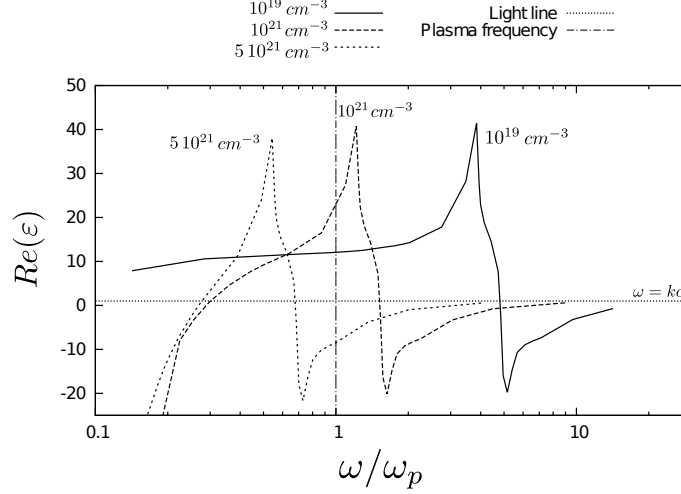


FIG. 1: Real part of the dielectric function of Si $\varepsilon(\omega) = \left(\frac{kc}{\omega}\right)^2$ as a function of the photon energy compared to the plasma frequency $\omega_p = \sqrt{\frac{n_e e^2}{m_e \varepsilon_0}}$. If the plasma frequency is located on the left of the resonance peak, surface mode is not bounded to the surface and SPP are not allowed. If the plasma frequency is located on the right of the surface plasmon polariton resonance peak, the SPPs are allowed.

(3) following the measurements of Palik⁴⁰. The wave-vector k is squared and normalized to the laser pulsation to obtain the dielectric function, and is presented as a function of the pulsation ω . The dispersion curve of the SPP reveals a plasmon resonance of the free-carrier plasma if the plasma frequency is comparable with the laser frequency e.g. if the free-carrier density is high. In the case of a doped semiconductor near the eigen frequency, the intersection of the dispersion curves is possible³¹. In the case of femtosecond laser interaction, similarly, laser-induced ionization provides free-carrier contribution to the dielectric function, which is equivalent to a transient doping. Therefore, the intersection also becomes possible. The bounding of the coupled wave to the surface is described by the condition $\omega < \frac{\omega_p}{\sqrt{2}}$ in a perfect conductor⁶⁵. In the case of damped material, however, the leaky part of the dispersion relation between $\omega_{spp} = \frac{\omega_p}{\sqrt{2}}$ and ω_p is allowed⁶⁴. The condition on pulsation in a damped material is thus described by

$$\omega < \omega_p. \quad (8)$$

The minimal density leading to satisfy this later condition is the critical density at which $\Re(\varepsilon) = 0$.

The coupling of far field laser wave with the SPP is also possible in the case of a surface

with defects or roughness³¹. In this case, the pseudo-grating period has a large thickness δk around its average value, and leads to a small but non-zero coupling efficiency. Because of the laser irradiation, a coupling of few percents is sufficient to obtain a periodic modulation of the deposited energy by interference between laser and SPP waves²⁸.

Finally, the conditions to satisfy (Eq. 5, Eq. 7, and Eq. 8) allowing the excitation of the SPP at the vacuum-Si interface can be combined into the condition

$$\Re(\varepsilon) < -1 \quad (9)$$

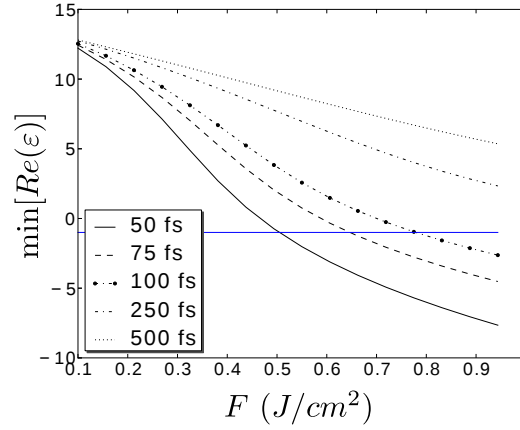
This criterion corresponds to a minimal free-carrier density given by $N_e = 4.27 \cdot 10^{21} \text{ cm}^{-3}$ in the case of a laser irradiation of Si using wavelength of $\lambda = 800 \text{ nm}$. Then, we determine laser parameters for which this condition is justified.

V. RESULTS AND DISCUSSION

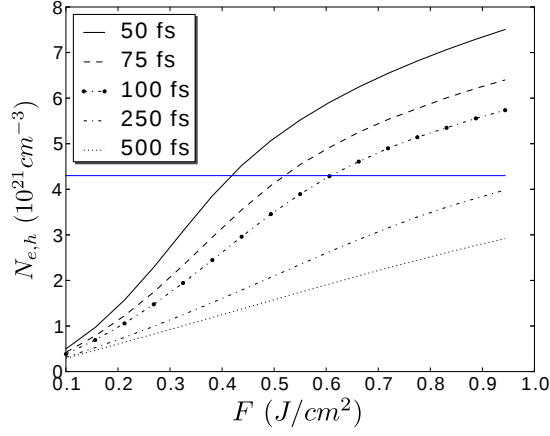
A. Conditions for SPP excitation on Si

We now demonstrate that excitation of Surface Plasmon Polariton is possible on Si under femtosecond laser irradiation, and calculate the laser parameters leading to satisfy the surface plasmon polariton conditions. Figure 2 (a) shows the maximum of the free-carrier density as a function of the laser fluence. At short pulse duration, the maximum free-carrier density is non-linear as a function of fluence. This effect is due to the modification of absorption near the critical density $n_{cr} = 3.98 \cdot 10^{21} \text{ cm}^{-3}$. At low laser intensity, the maximum density increases linearly, since the absorption is linear in this regime. Below critical density, one observe that the efficiency of the absorption increases, which is due to the significant contribution of the multi-photonic absorption. Above the critical density, absorption becomes limited by the surface reflectivity. Above this limit, the number of free carriers still increases due to high temperature of the free-carriers, leading to an interplay between diffusive transport and impact ionization.

Figure 2 (b) shows that the resonance condition is not met during the irradiation with a long pulse duration at the considered fluences, since the quantity of free-carriers is limited by the low intensity. It is shown that the real dielectric function decreases linearly if laser fluence is near the modification threshold (0.2 J/cm^2 , see Refs^{45,66,67}). We observe that the critical density is not reached in this fluence regime. The difference with Ref³⁶ is



(a)



(b)

FIG. 2: (a) Minimum of the real part of the dielectric function as a function of laser fluence. Blue line indicates the critical density. (b) Maximum of the density reached during the interaction as a function of laser fluence. Blue line indicates while $\Re(\varepsilon) = -1$. Several laser pulse durations are represented. Laser wavelength is 800 nm.

explained by the different collision time, the two-photon cross section which is 10 times lower here (see Ref⁶⁸), and the impact ionization that we took into account. In the case of 100 fs pulse duration, the condition given by Eq. (9) is satisfied above laser fluence of 0.7 J/cm^2 . A threshold for the excitation of SPP is then identified for a laser fluence of 0.7 J/cm^2 , a pulse duration of $\tau = 100 \text{ fs}$, and a laser wavelength of $\lambda = 800 \text{ nm}$. It is also shown that under shorter laser interaction, the condition for surface plasmon polariton excited is satisfied from lower fluences e.g. at 0.5 J/cm^2 if $\tau = 50 \text{ fs}$ and $\lambda = 800 \text{ nm}$.

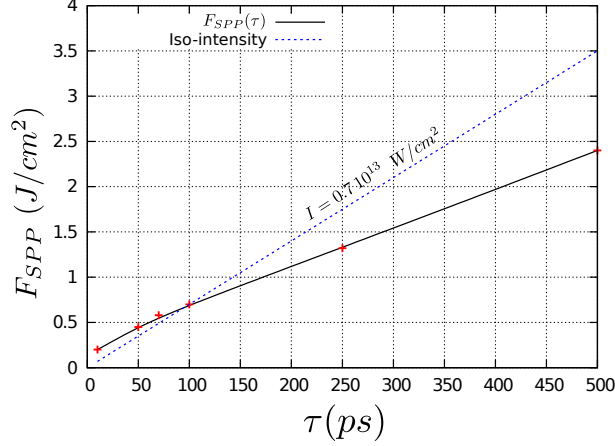


FIG. 3: Fluence threshold for SPP resonance as a function of pulse duration.

From those results, a threshold fluence for SPP resonance can be defined for each pulse duration, above which the SPP resonance conditions are met. This result explains why a high fluence is necessary to induce the formation of periodic structures in single pulse experiments^{4,26,66}, since it results in a sufficient quantity of free-carriers to excite surface waves at the surface of Silicon.

In Figure 3, we demonstrate the fluence threshold for the SPP resonance as a function of the laser pulse duration. The corresponding intensity, at which resonance occurs for 100 fs pulse, is shown by the dashed curve. By comparison of the curves, we observe that the required intensity for the SPP resonance increases with the decay in the laser pulse duration. This effect is due to the screening and large density gradient at the surface resulting into a strong diffusive transport.

Next we calculate the lifetime and the depth of the optically active zone, e.g. the distance under the surface where the sufficient number of free carriers are excited. Figure 4 shows the real part of the dielectric function as a function of time and depth. It is shown that under 50 fs pulse duration, at a fluence of 0.62 J/cm^2 , the excited zone is nearly 20 nm deep and the SPPs are excited during a picosecond, which is greater than the pulse duration, thus leading to the excitation of SPPs in a shorter timescale than necessary for surface melting. Moreover, the damping length of the SPP is given by the relation^{31,64} $L_{SPP} = [2\Im m(\beta)]^{-1}$. The value of the damping length is contained between 500 nm and $2 \mu\text{m}$ if the SPP resonance conditions are met. Then, the excited SPPs propagate through several micrometers and can lead to periodic modulation if laser interferes with the SPPs,

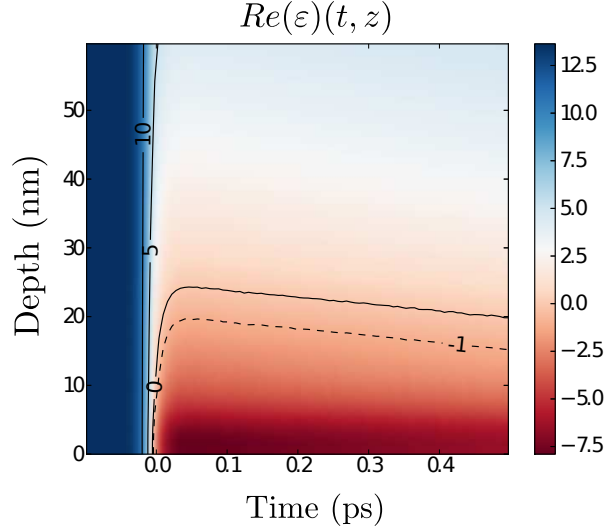
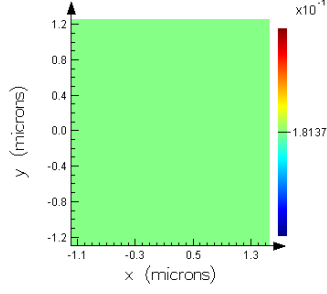


FIG. 4: Distribution of the dielectric function as a function of depth and time. $\tau = 50 \text{ fs}$,
 $F = 0.62 \text{ J/cm}^2$.

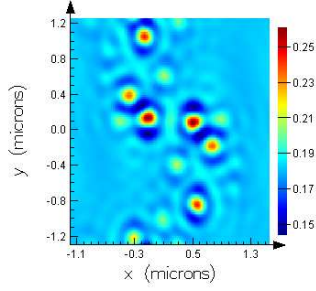
as experimentally observed around defects^{4,22,26,69,70}.

As underlined in the previous section, the phase-matching is possible at the surface of Si by using a scattering configuration with a defect or a roughness. We separate the following cases (i) the case of the roughness, for which the size of the scattering center is very small compared to the laser wavelength, and (ii) the case of a defect, for which the size is comparable to the laser wavelength. Both situations can lead to the excitation of Surface Plasmon Polariton if the conditions on the dielectric function are justified.

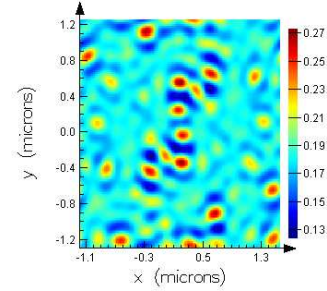
Figure 5 shows the distribution of the time-averaged transmitted field amplitude and normalized by the incident laser field intensity at the bottom of the selvedge region. This result has been calculated using the FDTD simulation package Lumerical⁷¹ by irradiating a randomly generated rough Si surface using several control parameters: FWHM of the amplitude, and distance between the scattering centers. In this calculation, the roughness amplitude is distributed as a gaussian function between 0 and 15 nm. The distance between scattering centers is taken equal to 100 nm so that the scattered waves interfere together. The Si dielectric constant is taken equal to the value under 800 nm wavelength laser irradiation, in the case of low laser excitation. One observes that the amplitude of the field transmitted below the roughness is modulated if roughness amplitude is greater than 8 nm. Such a roughness is formed after a single laser pulse⁷² at 0.5 J/cm^2 . Thus, the amplitude allowing the coupling of surface waves with laser is 8 nm, which explains why



(a) $\delta = 0$ nm



(b) $\delta = 8$ nm



(c) $\delta = 15$ nm

FIG. 5: Transmitted field amplitude below the selvedge region of Si. Roughness amplitude is (a) $\delta = 0$ nm, (b) $\delta = 8$ nm, (c) $\delta = 15$ nm at wavelength 800 nm. In these simulations, $\varepsilon = \varepsilon_\infty = 13.64 + 0.048i$.

strictly parallel ripples are observed after two pulses or more. Conversely, the formation of single pulse periodic structures is due to scattering on a near-wavelength defect, which leads to the excitation of Localized Surface Plasmon Polaritons distributed around scattering centers as observed by several authors^{22,26,69}. High fluence single pulse experiments leads to the formation of concentric structures rather oriented in the direction perpendicular to the laser polarization.

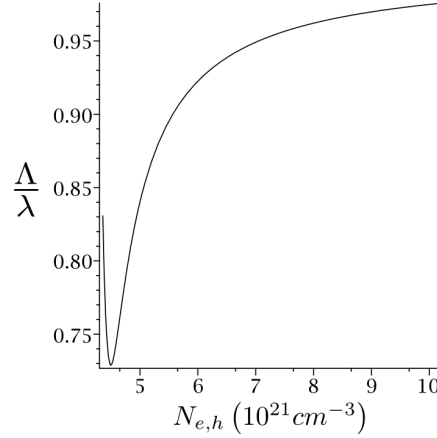


FIG. 6: Wavelength normalized period of SPP as a function of free-carrier density at vacuum - Si interface when the conditions of resonance are met.

In this section, we have theoretically demonstrated that the excitation of Surface Plasmon Polaritons occurs on Si irradiated by femtosecond lasers. The excitation conditions are satisfied during the laser pulse if the laser intensity is high. At 50 fs pulse duration, a layer of about 20 nm becomes optically active and has a lifetime longer than the pulse duration. We turn now to the study of the period of the SPPs excited during ultrashort laser pulse on Si.

B. Effect of the experimental parameters on SPP period

The SPP period dependency on laser intensity depends on the free-carrier density as follows

$$\Lambda = \frac{\lambda}{\sqrt{\frac{\varepsilon_1 \varepsilon_2(\omega)}{\varepsilon_1 + \varepsilon_2(\omega)}}}$$

where ε_1 and $\varepsilon_2(\omega)$ are respectively the dielectric functions of the media at both sides of the vacuum - Si interface. By substituting $\varepsilon_2(\omega)$ with Eq. (3), the periodicity of the SPP as a function of free-carrier density is calculated.

Figure 6 demonstrates the period of the SPPs at the vacuum - Si interface as a function of the free-carrier density. The values are presented for the free carrier number densities required for the SPP excitation. The resulting period varies considerably with the carrier

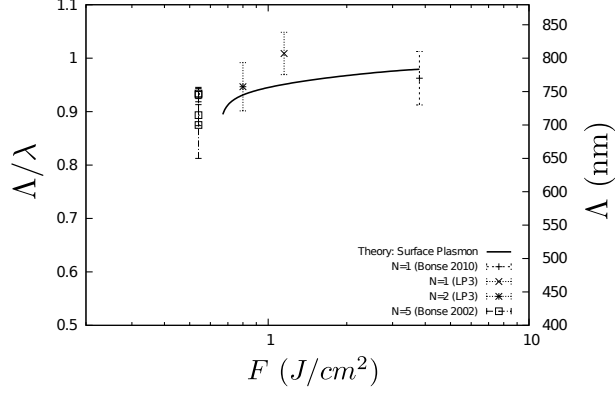


FIG. 7: Experimental measurements of the LSFL periods, as a function of laser fluence. $\tau = 100$ fs, $\lambda = 800$ nm, $\theta = 0^\circ$. The periods resulting from theoretical investigations are also represented. Bonse (2010) refers to Ref.⁴ and Bonse (2002) to Ref.⁶⁶

density. The period of the SPPs is contained between 0.7λ and λ , which correlates with the generally observed LSFL periodicities^{4,22,66}. A quantitative study of the variation of the SPP periodicity with laser fluence is now presented, and compared to the LSFL ripples formed using a very low number of laser pulses.

Figure 7 shows both theoretical and experimental periodicities. The period of the LSFL structures is presented as a function of laser fluence, for 100 fs pulse duration. When SPP resonance conditions are satisfied, the resulting SPP period tends to the laser wavelength when increasing laser fluence. In the optically active range (fluence is greater than $0.7 J/cm^2$ and pulse duration $\tau = 100$ fs), the calculation results agree with the presented experimental measurements taken from Refs^{4,66,72} at very low number of pulses. The single pulse case is explained by excitation of SPP via coupling with a surface defect. The case $N = 2$ is explained by coupling with roughness. This result shows that the periodicity of Surface Plasmon Polaritons well describes the evolution of the structure period as a function of the laser fluence at reduced pulse number.

In the case of 10 laser pulses, LSFL ripples are well-developed, and scattering by a grating model well explains the observed periodicities, which confirms Ref⁷³. Figure 8 shows the comparison between theoretical variation of period with angle of incidence with experiments made using 10 pulses at various fluences^{70,72}. Both directions of polarization are presented. The theory of scattering by a periodically structured surface leads to a

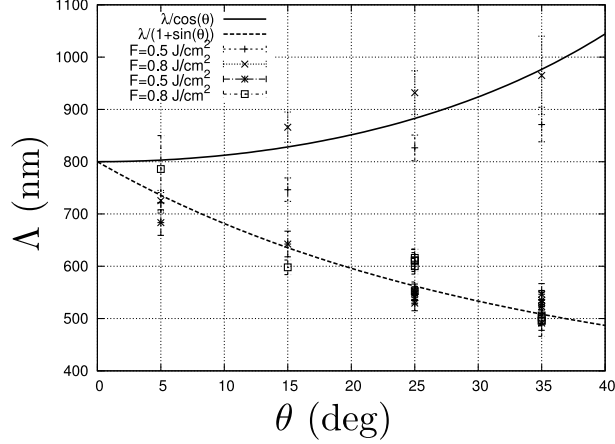


FIG. 8: Ripple periodicity as a function of angle of incidence and laser polarization after 10 laser pulses. Pulse duration is 100 fs, and laser wavelength is 800 nm.

periodicity given by⁶²

$$\Lambda_P = \frac{\lambda}{\sqrt{\eta^2 - \sin^2 \theta}}$$

where $\eta = \sqrt{\frac{\varepsilon_1 \varepsilon_2}{\varepsilon_1 + \varepsilon_2}}$. $\eta \sim 1$, $\Lambda_P \sim \frac{\lambda}{\cos \theta}$ for polarization parallel to the plane of incidence. For S polarization,

$$\Lambda_S = \frac{\lambda}{\eta + \sin \theta}$$

The variations of both measured and calculated ripple periods are explained by the variation of η near the critical density. Actually, $\eta \sim 1$ if $Re(\varepsilon) \ll -1$, and η varies between 0.5 and 1.5 if the condition (Eq. 9) is satisfied. This section demonstrated that the theory of SPP excitation on gratings agrees with the experiments and explains the variation of the ripple period with the angle of incidence and with laser polarization. We underline that the threshold fluence allowing SPP resonance on the surface of a grating is decreased with respect to the results presented in Figure 2 (a), since the energy absorption is enhanced in the presence of the grating⁶¹.

VI. CONCLUSIONS

The possibility of the surface plasmon polariton excitation on Si surface irradiated by a femtosecond laser pulse has been theoretically demonstrated. A sufficient number of free-carriers is excited from the valence band to the conduction band during the laser pulse, thus satisfying the SPP excitation conditions. The required ranges of laser fluences and

pulse durations have been identified to satisfy the SPP excitation conditions. In particular, SPPs can be excited by using a femtosecond laser with 800 nm wavelength, 100 fs pulse duration and with laser fluences larger than 0.7 J/cm^2 . The shorter temporal laser pulse width leads to the increase in the intensity threshold for the SPP excitation. This effect is due to the thermal diffusion of free-carriers and to the surface screening. As a result, a layer is excited, with a lifetime longer than the pulse duration and with a depth of several tens of nanometers.

Furthermore, a comparison of the calculated SPP periodicities and experimentally measured ripple periodicities allows us to conclude that the formation of periodic structures with a reduced number of laser pulses is due to the excitation of SPPs at the Si surface.

The presence of a surface roughness with $\delta \ll \lambda$ leads to the coupling of the laser wave with the roughness. We have found that the required roughness amplitude allowing the coupling of laser wave with the surface is 8 nm. It is also possible to obtain periodic structures by scattering on defects ($\delta \sim \lambda$) that are present at the surface by excitation of localized surface plasmon polaritons. These results underline the importance of the surface quality in the SPP excitation and thus to the LSFL ripple formation.

As a result of the performed analysis, the possibilities of control over the period of the LSFL ripples can be deduced in the regime of low number of pulses. The period can be reduced down to 40% by increasing the angle of incidence for S polarization, and can be increased up to 37% by increasing the angle of incidence for P polarization. The period of the LSFL ripples can be increased by about 10% by increasing the laser fluence up to 5 J/cm^2 . Finally, one should mention that LSFL period reduction due with pulse number also allows to decrease the periodicity of 50%, but the corresponding mechanism is still under discussions^{4,25}.

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